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## Report Title

An Optical Lattice Clock with Spin-1/2 Atoms

### ABSTRACT

An optical lattice clock probes a spectrally narrow electronic transition in an ensemble of optically trapped, laser-cooled atoms, for use as a time and frequency standard. To date, several lattice clocks have been demonstrated with superior stability and accuracy compared to primary frequency standards based on microwave transitions. Yet, the question of which atomic system (including the element and isotope) performs best as a lattice clock remains unsettled. This thesis describes the first detailed investigation of an optical lattice clock using a spin-1/2 isotope of the ytterbium atom. A spin-1/2 system possesses several advantages over higher-spin systems, including a simplified level structure (allowing for straightforward manipulation of the nuclear spin state) and absence of any tensor light shift from the confining optical lattice. Moreover, the ytterbium atom (Yb) stands among the leading lattice clock candidates, offering a high performance optical clock with some degree of experimental simplicity. The frequency stability of the Yb clock is highlighted by resolving an ultra-narrow clock spectrum with a full-width at half maximum of 1 Hz, corresponding to a record quality factor  $Q = \nu/\Delta\nu = 5 \times 10^{14}$ . Moreover, this system can be highly accurate, which is demonstrated by characterizing the Yb clock frequency at the  $3 \times 10^{-16}$  level of fractional uncertainty, with further progress toward a ten-fold improvement also presented. To reach this low level of uncertainty required careful consideration of important systematic errors, including the identification of the Stark-canceling wavelength, where the clock's sensitivity to the lattice intensity is minimized, a precise determination of the static polarizability of the clock transition, and the measurement and control of the atom-atom collisions.

# Optical Lattice Clock with Spin-1/2 Ytterbium Atoms

by

**N. D. Lemke**

B.S., Bethel University, 2006

A thesis submitted to the  
Faculty of the Graduate School of the  
University of Colorado in partial fulfillment  
of the requirements for the degree of  
Doctor of Philosophy  
Department of Physics

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Lemke, N. D. (Ph.D., Physics)

Optical Lattice Clock with Spin-1/2 Ytterbium Atoms

Thesis directed by Dr. Jun Ye

An optical lattice clock probes a spectrally narrow electronic transition in an ensemble of optically trapped, laser-cooled atoms, for use as a time and frequency standard. To date, several lattice clocks have been demonstrated with superior stability and accuracy compared to primary frequency standards based on microwave transitions. Yet, the question of which atomic system (including the element and isotope) performs best as a lattice clock remains unsettled. This thesis describes the first detailed investigation of an optical lattice clock using a spin-1/2 isotope of the ytterbium atom. A spin-1/2 system possesses several advantages over higher-spin systems, including a simplified level structure (allowing for straightforward manipulation of the nuclear spin state) and the absence of any tensor light shift from the confining optical lattice. Moreover, the ytterbium atom (Yb) stands among the leading lattice clock candidates, offering a high-performance optical clock with some degree of experimental simplicity. The frequency stability of the Yb clock is highlighted by resolving an ultra-narrow clock spectrum with a full-width at half-maximum of 1 Hz, corresponding to a record quality factor  $Q = \nu_0/\Delta\nu = 5 \times 10^{14}$ . Moreover, this system can be highly accurate, which is demonstrated by characterizing the Yb clock frequency at the  $3 \times 10^{-16}$  level of fractional uncertainty, with further progress toward a ten-fold improvement also presented. To reach this low level of uncertainty required careful consideration of important systematic errors, including the identification of the Stark-canceling wavelength, where the clock's sensitivity to the lattice intensity is minimized, a precise determination of the static polarizability of the clock transition, and the measurement and control of atom-atom collisions.

## Dedication

To my family.

PREVIEW

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# Chapter 1

## Introduction

This thesis details the first experimental investigation of an optical lattice clock using an isotope with nuclear spin  $I = 1/2$ . While lattice clocks based on atoms with higher nuclear spin [1] and with no nuclear spin [2] have been demonstrated, here we highlight a few special properties of a spin-1/2 system. To set the stage for this discussion, we will first review some basic principles of atomic clocks, followed by a brief discussion of several applications that call for higher-performing atomic clocks. Then, in Sec. 1.4, we will examine why a spin-1/2 system may be the best choice for state-of-the-art timekeeping with an optical lattice clock.

### 1.1 Atomic clocks

Since 1967, the unit of time in the International System of Units (SI) has been defined in terms of the energy difference between the two lowest states of cesium atoms [3, 4]<sup>1</sup>. This definition is realized in the laboratory by steering a microwave oscillator to stay resonant with the Cs atoms while simultaneously counting the number of oscillations (“ticks”) that have passed. Atomic transitions are ideally suited for timekeeping because the atom’s energy levels are largely immune to environmental effects. Because microwave radiation oscillates very quickly (about 10 billion times per second), each cycle is short in time, allowing for highly precise measurements of time and frequency. Today’s Cs primary frequency standards are extremely accurate, with uncertainty at just a few parts in  $10^{16}$  [5, 6, 7, 8, 9, 10]. This is equivalent to saying that the clock

---

<sup>1</sup> Specifically, the definition of the second is “the duration of 9 192 631 770 periods of the radiation corresponding to the transition between the two hyperfine levels of the ground state of the caesium-133 atom” [3].

would neither gain nor lose one second in 100 million years.

This exceptional accuracy is made possible by the stability, or precision, of atomic clocks. Two figures of merit that largely determine the stability of the clock are its resonance quality factor ( $Q = \nu_0/\Delta\nu$ ) and its measurement signal-to-noise ratio (S/N). The quality factor is a dimensionless quantity that gives the width of a resonance,  $\Delta\nu$ , in comparison to its center frequency,  $\nu_0$ . For the best Cs clocks, a linewidth of 1 Hz results in  $Q = 10^{10}$ . The S/N is also dimensionless, and it gives information about the fidelity of each measurement. In general, there are many sources of noise that could diminish this quantity, but under certain conditions it is possible to reach the “standard quantum limit” of  $S/N = \sqrt{N_{atom}}$  with  $N_{atom}$  the number of atoms interrogated. For a typical value of  $10^6$  atoms, the S/N is 1000. The product of  $Q$  and S/N gives the fractional precision (here,  $10^{13}$ ), which is indeed not far from the typical stability of Cs fountain clocks [5, 9]. This means that after just one measurement (lasting approximately 1 second), the frequency has already been found to 13 digits, and subsequent measurements strung together will continue to increase the combined precision. With this level of stability, it is possible to quickly and thoroughly evaluate possible sources of systematic error and to compare two clocks to see how well they agree.

To achieve this level of inaccuracy has taken more than 50 years of scientific and technical advances. One very important step along the way was the advent of laser cooling of atoms [11, 12] and its application to atomic clocks [13, 14]. The key advantage to cold atoms lies in their slowed velocity, which results in longer interaction times and reduced Doppler shifts. State-of-the-art microwave clocks typically use a fountain geometry, in which a ball of laser-cooled atoms is tossed straight up before falling back under gravity. This atomic fountain, which is very successful at canceling Doppler effects [15], is made possible by the ultracold atomic temperature.

So what limits the accuracy of cold-atom microwave fountain clocks such as NIST-F1? The full answer includes a long list of systematic effects, each of which needs to be evaluated to smaller and smaller levels of uncertainty. But with a fixed  $Q$  and a clock stability approaching its fundamental limit, smaller uncertainties require longer averaging times, and eventually this becomes impractical or even impossible. For this reason, it is interesting to consider atomic systems with

potentially higher stability, which in turn offers the possibility for a corresponding reduction in absolute uncertainty.

## 1.2 Optical clocks and the optical frequency comb

The clearest path for developing a higher stability clock is to use a higher frequency transition. Optical transitions (so-called because they absorb and emit light visible to human eyes) have frequencies in the hundreds of terahertz range, or nearly 100 000 times higher than microwave clocks. These optical clocks thus offer tremendous advantages for improved time and frequency metrology, and sometime (perhaps in the next 10 years) the SI second will be redefined in terms of an optical clock transition.

In the past, one disadvantage to optical frequency standards was the lack of an optical clockwork — a way of counting the ticks [16, 17, 18, 19, 20]. Because optical frequencies are so high ( $10^{15}$  oscillations per second), standard electronic counters cannot be used. Around the year 2000, this problem was solved by the development of the optical frequency comb, for which a portion of the 2005 Nobel Prize in physics was awarded [21, 22, 23, 24, 25, 26, 27, 28]. The optical frequency comb is a pulsed laser that outputs very short pulses of light. Each pulse is composed of thousands of laser modes that form a grid (or a “comb”) in frequency space, much like the fine tick marks on a ruler that allow for precise length measurements (see Fig. 1.1(a)). These optical frequency combs are useful both for comparing two optical clocks at different frequencies and for dividing an optical signal to the microwave domain where it can be counted, and they have enabled vast progress in the field of optical frequency metrology over the last decade. We will consider the comb in slightly more detail in Ch. 7.

The basic scheme of an optical clock is shown in Fig 1.1(b). A laser is electronically stabilized to a high-finesse optical cavity, and the cavity-locked laser comprises the clock’s oscillator (typically called the local oscillator, or LO). The optical cavity is designed to have a very constant length (i.e., the distance between the two reflecting mirrors), as the length stability of the cavity directly determines the frequency stability of the laser. While the optical cavity provides an excellent means

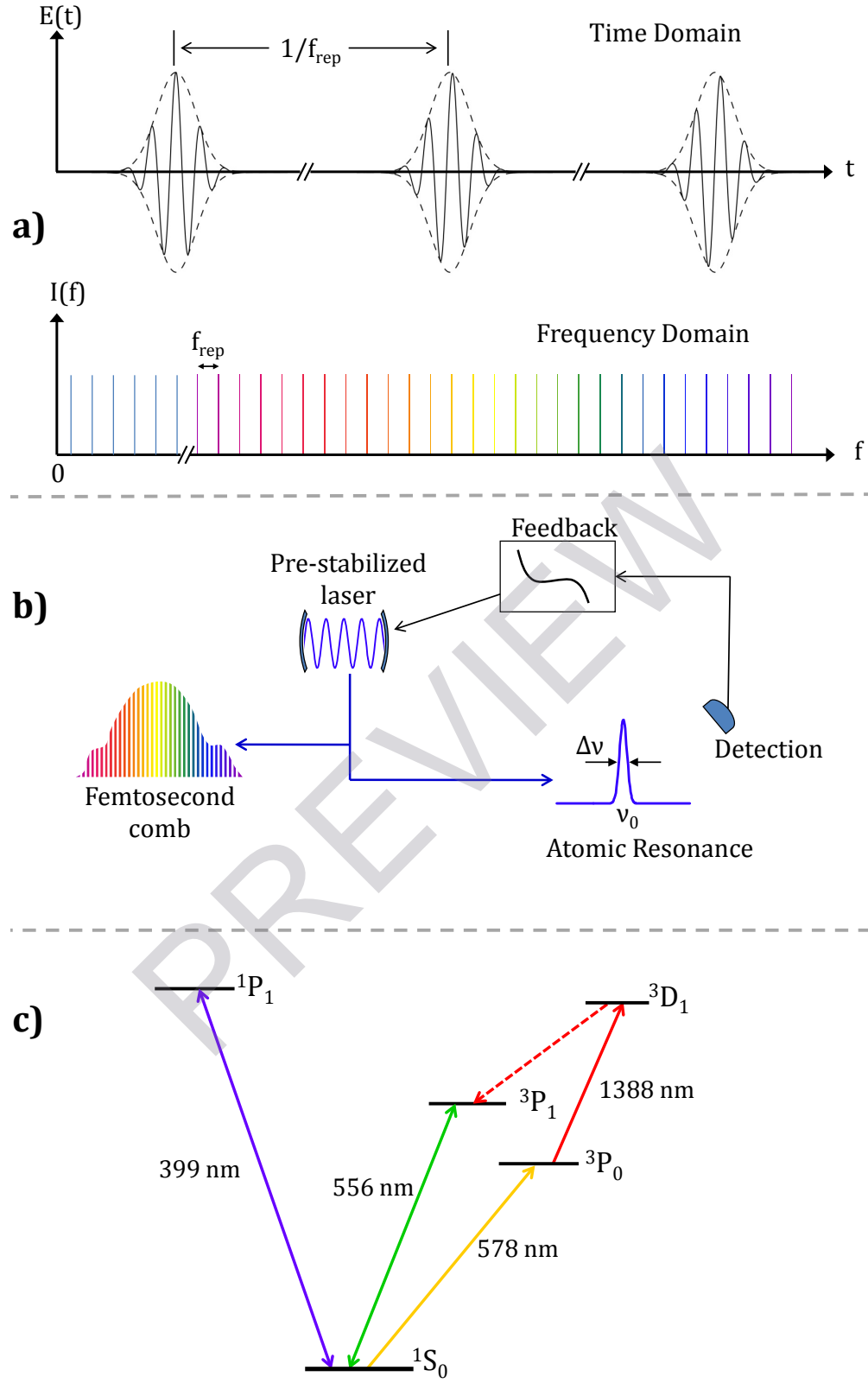


Figure 1.1: a) Time- and frequency-domain depiction of the frequency comb output. b) Block diagram of an optical clock. c) Relevant energy levels in Yb.

for reducing the noise of the laser, it does not provide a good long-term reference (nor is its length a fundamental physical quantity with which to form a universal standard), so an atomic system serves as the long-term frequency reference. Specifically, light from the cavity-stabilized laser probes an optical transition in the atomic system, and electronic corrections are applied to the laser frequency to hold it on resonance with the atom(s). The final component in the optical clock scheme is the optical frequency comb, as mentioned above, which acts as a frequency counter by accumulating trillions of oscillations on the optical signal before outputting a “tick”.

Two competing technologies for the atomic reference are currently being pursued at metrology labs throughout the world: trapped-ion clocks and optical lattice clocks [29, 30]. While there are a number of factors that will play into the ultimate decision of which atomic system becomes the new standard, not to mention which system(s) will perform best as a portable device for commercial and space applications, it is likely that both types will find useful applications. Let us first examine the potential stability of these systems. Trapped-ion clocks typically interrogate one electrically-charged atom, confined by RF electromagnetic fields and cooled to its motional ground state [31, 32, 33, 34, 35, 36, 37]. These ion clocks have thus far been limited to one clock atom because of the possible clock errors due to Coulomb repulsion between multiple atoms. For a single-trapped-ion clock, atomic spectra of few-hertz width [38, 39] and a S/N of 1 yield a fractional precision of  $10^{14}$  in a single measurement.

Optical lattice clocks, by contrast, interrogate an ensemble of laser-cooled atoms confined in an optical potential [40, 41, 42, 1, 43]. This optical potential is formed by a standing-wave laser field known as an optical lattice and is tuned to a specific wavelength (the “magic wavelength”) where it does not perturb the clock frequency [40, 44]. Just as with ion clocks, the resonances in lattice clocks can be very narrow, leading to similar  $Q$ -factors of  $10^{14}$  or higher [45, 43, 46]. But, here the large number of quantum absorbers results in a S/N of 100 to 1000, leading to a potential precision exceeding  $10^{17}$ . From these considerations, we can see the optical lattice clocks have the potential for 10,000-fold improvement in stability over microwave clocks. Moreover, as we will see later in this thesis, these systems also possess the potential to be highly accurate, making them excellent

candidates for state-of-the-art metrology. However, we also note that many of these gains have yet to be realized. Due to technical noise sources, the stability of lattice clocks has not reached the level mentioned above, but is instead similar to that of the ion clocks [47, 1, 48, 46]. Moreover, the best accuracy demonstrated so far [1, 49, 43, 50] is only slightly better than that of the best microwave clocks, and significantly worse than that of the best ion clocks [36, 37]. So while these lattice clock systems remain tantalizing, there are still many significant obstacles to overcome. In this thesis, we will examine some of these obstacles and demonstrate some new techniques to overcome them.

### 1.3 Applications for ultra-precise clocks

Many of today’s technologies in communications and navigation (notably GPS) have been enabled by atomic clocks, and it is expected that the vast improvements in timekeeping offered by optical clocks will further enable new technologies. While it is impossible to fully predict what scientific and technological impact optical clocks will ultimately have, we can already list some known applications for these higher-performing clocks. Following are a few of the most significant.

#### i) Variation of constants

The search for time-variation in the fundamental constants is an active field of research in which atomic clocks are just one piece, albeit an important one. There are several reasons to search for such variations, ranging from tests of new cosmological and unification theories to explanations for the “fine-tuning” question of why the current values of fundamental constants are capable of supporting life on earth [51, 52, 53]. While astronomical data are useful for assessing the values of fundamental constants in the distant past, laboratory searches offer precise measurements of the current drift rates in a highly controlled setting. Unlike microwave transitions, which are primarily sensitive to changes in the electron-proton mass ratio ( $\mu = m_e/m_p$ ), optical transitions are primarily sensitive to changes in the fine structure constant,  $\alpha = e^2/(4\pi\epsilon_0\hbar c)$  [54, 55]. In fact, the tightest constraint to date on  $\dot{\alpha}$  was set by the  $\text{Al}^+$  and  $\text{Hg}^+$  optical clocks at NIST [36]. Fractional changes in the clock transition frequency are

related to changes in  $\alpha$  by

$$\frac{\delta\nu}{\nu} = K \frac{\delta\alpha}{\alpha} \quad (1.1)$$

where  $K$  depends on the atomic structure. Generally, lattice clocks do not have especially high sensitivities ( $K_{\text{Sr}} = 0.06$ ,  $K_{\text{Yb}} = 0.32$ ,  $K_{\text{Hg}^+} = -2.94$ ) so they may be most useful in this regard by serving as “anchor” lines against which atomic systems with higher sensitivity may be compared [56]. In the same manner, the Sr lattice clock transition, which has been compared to the Cs microwave transition in several labs throughout the world, served as a stable reference for constraining  $\dot{\mu}$  in Cs [55].

ii) Testing relativity and the equivalence principle

Local position invariance (LPI), which is a consequence of Einstein’s equivalence principle [57, 58], requires that the outcome of a non-gravitational measurement does not depend on the value of the local gravitational potential. Space-born optical clocks could test this by measuring the frequency ratio of two clocks of different species on a satellite and comparing it to the value measured on earth. Alternatively, the ratio could be monitored as the satellite traverses a highly eccentric orbit, thus modulating the gravitational potential in time. In this case the constraints on the two clocks’ accuracies are reduced, though their medium-term (few hour) stability must be quite good. These techniques are expected to improve upon the best measurement of LPI by more than 5 orders of magnitude [59, 60].

Similarly, the absolute gravitational redshift could be measured with space-based clocks, either with two clocks on different circular orbits, or with one clock in a highly-eccentric orbit and one stationary [59]. Here the expected improvement from previous results is four orders [60]. Additionally, with a reference clock in space, earth’s gravitational field could be measured with greater accuracy, enabling an improved mapping of the geoid [61].

Some unification theories predict that the values of fundamental constants are coupled to the gravitational field, which would violate LPI. While this could be explored most neatly with a deep space mission, it could also be observable on earth due to the ellipticity of earth’s orbit.



Any absence of change in the clock frequency, over the timescale of earth's orbital period, constrains the possible size of these couplings [55].

iii) Low noise microwave generation

Photodetection of a femtosecond frequency comb, referenced to an optical clock, yields an electronic signal at a harmonic of the repetition rate (typically chosen near 10 GHz) with exceptionally low phase noise. While there are additional noise sources that can play a role, these are tractable, and microwave signals whose noise properties are nearly as good as those of the optical clock have been produced [62, 63]. The technical applications calling for these low phase noise microwaves include the following: remote synchronization of large scientific facilities such as synchrotron and accelerators [64, 65]; the local oscillator in a microwave clock [66]; and very-long baseline interferometry (VLBI), in which signals at spatially separate radio telescopes can be combined (with the help of a fine timing system) to yield exceptional angular resolution in studying astrophysical phenomena [67].

iv) Realization of SI units

As mentioned above, the SI second is defined by a transition in atomic cesium, requiring Cs atomic clocks to realize the definition. While some optical clocks have already demonstrated superior repeatability compared to the best microwave clocks, they cannot, by definition, be accurate. For this reason, the definition of the SI second will almost certainly be changed someday to an optical transition, which will then require metrology labs the world over to build new atomic clocks based on the particular optical transition selected.

With the speed of light defined to be exactly  $c = 299\,792\,458$  m/s, the meter is then defined as the distance light travels (in vacuum) in a time of  $1/c \approx 3$  ns [68]. This definition can only be realized accurately with a Cs atomic clock, which defines the second, together with an optical frequency comb that links visible laser light with the microwave clock. However, some optical transitions, including those of  $\text{Hg}^+$  ion and Sr lattice, have been recognized as secondary representations of the SI second, which means they also can be used for realizing

the SI meter. The accuracy gained by defining the second by an optical transition could, at least in principle, lead to more accurate length measurements as well. With better measures of the fine structure and Rydberg constants, it may be possible to someday realize other physical quantities (like mass) with optical atomic clocks [69].

#### 1.4 Spin-1/2 ytterbium atoms

The first proposal for an optical lattice clock called for spectroscopy of a narrow optical transition in ultracold strontium atoms [40]. Since then, experimental groups have begun researching not only strontium (Sr) [70, 71, 72, 73, 74], but also ytterbium (Yb) [75, 76, 77, 78, 79] and mercury (Hg) [80, 81] as lattice clock candidates. Because the electronic structure is very similar for all atoms with two valence electrons, the lattice clock scheme could in principle be employed with Be, Mg, Ca, Ba, Zn, Cd, Ra, and the synthetic element nobelium (No).

In these divalent atoms, the electronic structure can be arranged by symmetry into singlet states and triplet states, and transitions between the two manifolds are generally forbidden. The two clock states are the  $^1S_0$  ground state and the  $^3P_0$  excited state, and the radiative transition between them is both spin- and dipole-forbidden. This gives the excited state a very long lifetime, and potentially allows for the observation of an extremely narrow clock transition, which yields a large  $Q$ -factor as discussed above. In fact, in isotopes lacking nuclear spin, the  $^3P_0$  lifetime has been calculated to be a few thousand years [82]! While a long-lived excited state is an essential ingredient in the lattice clock scheme, this is actually far too long to be useful, because the transition is too weak to efficiently excite.

For this reason, most experiments use an isotope that does have nuclear spin. Here, the hyperfine interaction perturbs the excited clock state ( $^3P_0$ ), mixing it with other nearby states and typically resulting in a hyperfine-quenched  $^3P_0$  lifetime of several tens of seconds [83, 84]. These odd-mass-number isotopes are less abundant than the (spin-0) even-mass-number isotopes, but usually the odd isotopes have sufficient abundance to carry out experiments.<sup>2</sup> The amount of

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<sup>2</sup> One notable exception is Ca, in which the only odd isotope (43) has 0.135 % abundance.